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ENVIRONMENTAL HEALTH SERIES

Radiological Health

AN ACCLIMATION ROOM FOR THE DETECTION OF LOW RADIUM 226 BODY BURDENS

U. S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE

Public Health Service

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AN ACCLIMATION ROOM FOR THE DETECTION OF LOW RADIUM 226 BODY BURDENS

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U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE

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ABSTRACT

A mobile breath radon detection apparatus has been used for measuring low radium 226 body burdens. The unique feature of the apparatus is an acclimation room designed to accommodate subjects for periods up to 1 week in a low-radon environment. Radon concentrations in the acclimation room have been reduced to an operating level of 0.0005 pCi per liter, a reduction of approximately 600:1, by an air processing unit employing activated coconut charcoal. This apparatus permits determination of very low radium 226 body burdens in comfortable surroundings for two subjects simultaneously, during the extended periods required for elimination of previously inhaled radon from the body. The construction and operation of the acclimation room and breath sample collection are described. Radium 226 body burdens for the subjects and a radon elimination curve for one subject are included.

Representative products and manufacturers are named for identification only, and listing does not imply endorsement by the Public Health Service and the U.S. Department of Health, Education, and Welfare.

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An Acclimation Room for the Detection of Low Radium 226 Body Burdens

INTRODUCTION

The breath radon method for determining radium 226 body burdens resulting from therapeutic or occupational exposure has been in use for half a century (*1*). Rajewsky *et al.* have reviewed the development of breath radon devices used for determining the radium body burden in man (*2*). Early investigators, limited by the sensitivity of their instruments, were unable to detect radon 222 concentrations below those corresponding to the maximum permissible radium 226 body burden for occupational exposure (0.1 μg). Improvements in instrumentation have since resulted in the detection of body burdens several orders of magnitude below those previously measured.

Retention of inhaled atmospheric radon by soft tissue in the body, changes in respiratory activity, and sample contamination by radon from breath collection equipment have been obstacles to further refinement of breath radon techniques for determining radium 226 body burdens (*3–5*). Harley *et al.* and Stehney *et al.* have identified retention of inhaled radon in the soft tissue as a major interfering factor (*3, 6*). They have shown that breathing "radon-free" air for an extended period of time is required to significantly reduce the radon concentration in the breath caused by body retention of inhaled atmospheric radon. In their studies, radon in the breath resulting from inhalation of atmospheric radon was reduced by administering aged air to the subjects through a face mask or hood. In the study by Stehney *et al.* subjects breathed aged air for 12 hours prior to the breath sampling.

A system was developed to permit determination of low radium 226 body burdens without requiring the subjects to wear face

masks or hoods for extended periods. Its unique feature is an acclimation room designed to accommodate subjects for periods up to 1 week in a low-radon environment. The apparatus was developed as a part of the U.S. Public Health Service San Juan Basin Radium Research Project for the determination of radium 226 body burdens in a population with an environmental radium 226 intake of approximately 30 pCi per day (an intake slightly in excess of that corresponding to the Federal Radiation Council's Radiation Protection Guide (RPG) of 0.001 μCi radium 226 average adult skeletal body burden in an exposed population group (*7*)).

It was expected that subjects might have to remain in a low-radon atmosphere for periods exceeding 1 day prior to breath sampling because of the relatively low radium 226 body burdens anticipated. Acclimation in a low-radon atmosphere was considered necessary to eliminate sufficient amounts of atmospheric radon from the body in order to detect accurately the relatively small amounts of radon emanating from the low radium 226 body burdens. Since measurements were to be made on individuals who might be reluctant to wear a face mask or hood for extended periods, the controlled-atmosphere room was developed to provide low-radon air in more comfortable surroundings.

DESIGN CONSIDERATIONS

Design considerations were based on the achievement of a low-radon atmosphere in the acclimation room, an environment conducive to subject cooperation, and mobility of the entire system.

The first step in achieving a low-radon atmosphere was to determine the atmospheric radon level that would not obscure the radon resulting from radioactive decay of radium 226 in the body. A 100 pCi radium 226 body burden would produce a radon 222 concentration in the breath of 0.88 fCi per liter (assuming a breathing rate of 10 liters per minute and exhalation of 70 percent of the radon formed). The average atmospheric radon concentration at the laboratory site was found to be 266 fCi per liter. Over 99 percent of this radon had to be removed to reduce the atmospheric radon concentration to the same order of magnitude as that produced in the breath by a 100 pCi radium 226

body burden; therefore the air processing unit was designed to reduce radon concentrations in the acclimation room by a factor of at least 100 below the radon concentration of outside air.

It is insufficient to provide air of low-radon content only during the period of sample collection. Harley *et al.*, using elevated radon concentrations, have shown that approximately 15 hours of breathing radon-free air is required to reduce the radon levels in the breath to one one-thousandths of the concentration in the previously inspired air (*3*). Figure 1, which extrapolates data from Harley *et al.* (*3*), shows the relative contribution of radon in the breath from different sources as a function of the subject's time in the acclimation room. In this example, after 12 to 15 hours acclimation, radon resulting from previously inspired air would be reduced to a small fraction of the total breath radon.

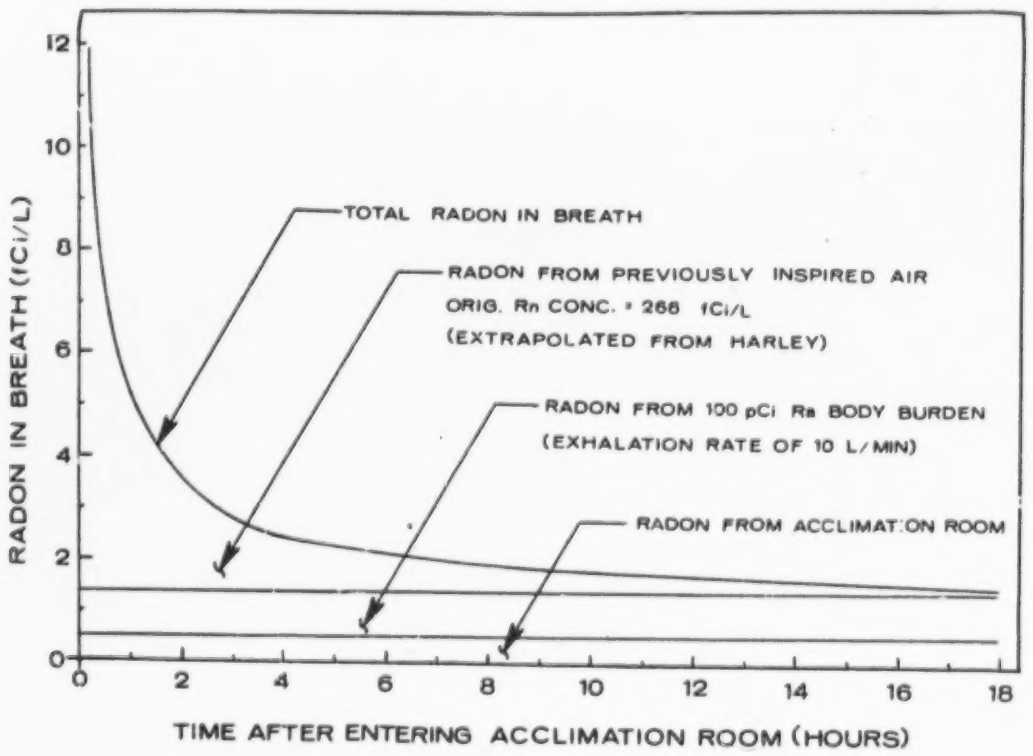

Figure 1. Sources of radon in breath (theoretical)

It should be emphasized that the main difficulty in determining radium body burdens is not in the detectable limits of radon, instead it is in ascertaining the true contribution of radium in the body to the concentration of radon in the breath. The amount of radon in the breath resulting from the previously inspired air can only be estimated. This represents a source of error which can be minimized by a long acclimation period. While radon in

the breath resulting from radon in the air of the acclimation room can be measured, it should also be minimized. Therefore, it was considered necessary not only to remove radon from the room air, but also to identify and limit equipment, building materials, and furnishings containing small amounts of radium which could contribute significantly to higher radon concentrations in the air and subsequently the breath.

In order to obtain subject cooperation in this study, the acclimation room was designed, and sampling procedures were developed to provide as much comfort as possible for testing periods up to 1 week. The acclimation room was designed to accommodate two adult subjects simultaneously.

Mobility of the apparatus was desirable, both for subject convenience and for flexibility of use. Thus all of the systems were either built into trailers or constructed in such a fashion so they could be transported and operated with a minimum of supporting facilities.

APPARATUS

The breath radon apparatus was divided into five separate but integrated systems: the acclimation room, air processing unit, collection system, radon transfer system, and counting equipment. To provide mobility, the apparatus was divided into three components. The acclimation room, collection system, and food preparation center were all mounted in one 20-foot trailer. The transfer system, counting system, and associated laboratory equipment were housed in an adjacent trailer. The air processing unit was skid-mounted so it could be transported on a flat-bed trailer.

Acclimation Room

The construction material used in the acclimation room was specially selected for its low-radon emanation rate. The room was steel-lined and painted with deck gray and apple-green water-

based paint. Several paints were tested for possible radon emission by hanging 10 foot by 1 foot strips of aluminum foil, painted on both sides, in the room overnight. Radon backgrounds were measured before the painted foils were placed in the room and again the next morning. Successive radon backgrounds were also measured as each piece of furniture was added to the room. This selection process helped to insure minimum radon contributions to the processed air.

The interior of the room, 10 feet by 7 feet by 6 feet, is shown in figure 2. Within the room were two bunk beds, wash basin, television set, radio, telephone, table and chairs, a chemical toilet, curtained windows, and a comfort air conditioner. Safety features included an intercom system to the operator on watch, and an emergency window exit.

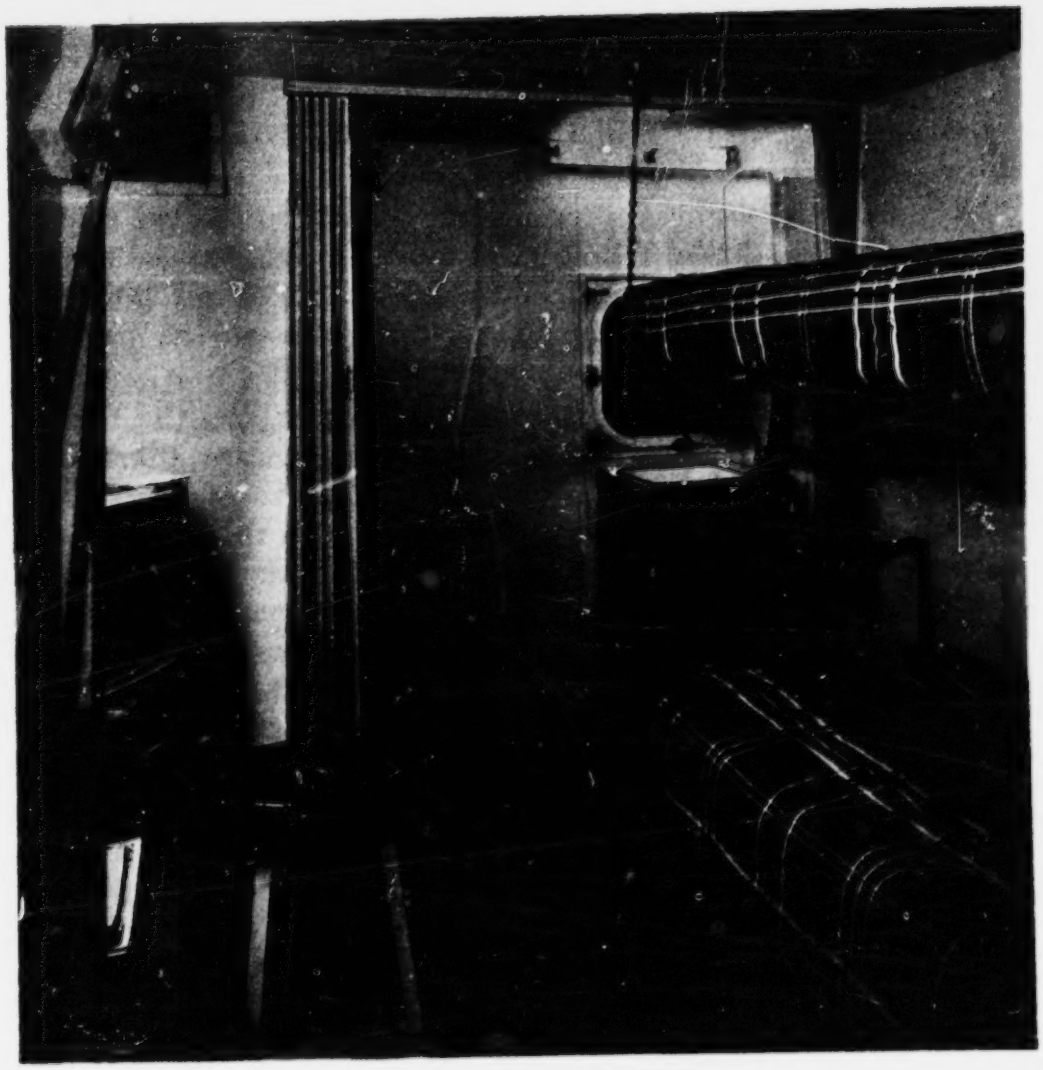

Figure 2. Acclimation room

The air supply to the acclimation room consisted of 55 SCFM of air, which was processed for moisture, carbon dioxide, and radon removal. Inside the room the air was maintained at a positive pressure of ¼ to ½ inch of water with respect to the outside atmosphere. Thus any leaks would be outward from the acclimation room.

Air Processing Unit

The air processing unit was designed to reduce the radon concentration in the acclimation room to a level below that produced in the breath by a low-radium body burden. The unit consisted of an air compressor, refrigeration units, moisture condenser, drying unit, radon absorption towers containing activated charcoal, electric heaters, and ultra filters (fig. 3). The unit was 20 feet long, 8 feet wide, 8 feet high, and weighed 7 tons.

All piping and fixtures were copper, joined with silver solder, or stainless steel, threaded and teflon taped. A water-sealed rotary pump was used to provide a continuous nonpulsed air flow. No chemicals that contained calcium or members of the calcium family greater than in-trace amounts were used.

Prior to startup of the air circulation equipment, the towers containing the activated coconut charcoal beds were alternately evacuated and flushed with aged nitrogen to remove air. The charcoal was then heated to at least 500° F. with thermostatically controlled electric heaters, evacuated, and then flushed again with aged nitrogen. This purged any radon from the charcoal. The three charcoal towers were designed so they could be used singly, in series, or in parallel. It was possible also to regenerate one tower while operating another. For these studies, a single tower was used at one time since the acclimation room radon concentration could be kept sufficiently low for more than 24 hours by this procedure.

The heating portion of the regeneration process required a minimum of 2 hours, after which the tower was allowed to cool to a point so that none of the three thermocouples, located in the bed, indicated a temperature greater than 180° F. At this indicated temperature, air could be passed through the charcoal without danger of ignition or CO production. During normal op-

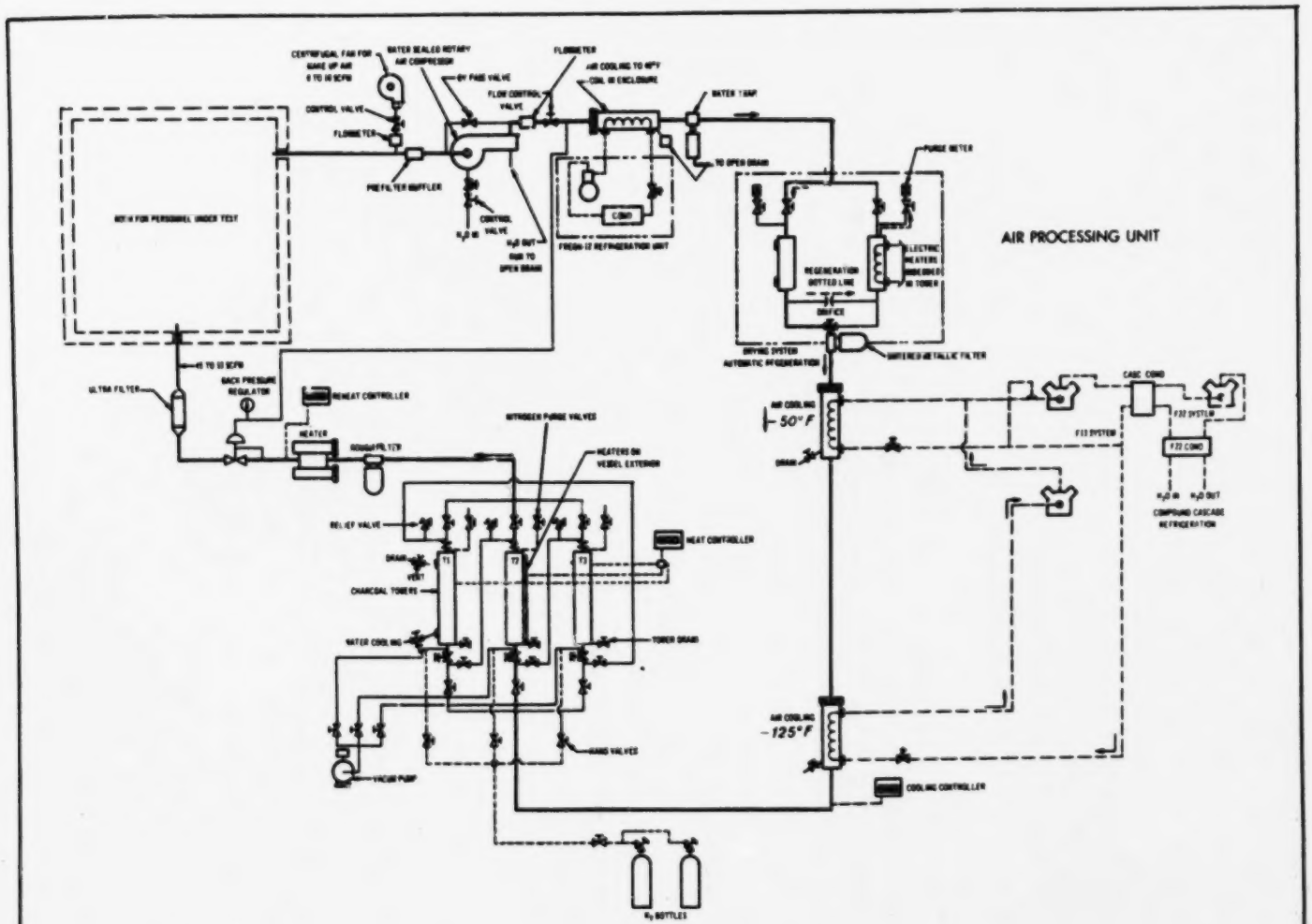

Figure 3. Flow diagram for air processing unit

eration the charcoal bed was regenerated 1 day prior to use, thus allowing time for the bed to cool from 180° F. to room temperature.

After regeneration, the system was ready to begin processing air. Air, at room temperature, left the acclimation room at 45 SCFM and was blended with makeup air at 10 SCFM from a centrifugal fan, throttling valve, and rotometer. The 55 SCFM of air was passed through a muffler (to reduce compressor noise) and then compressed by a water-sealed rotary air compressor to 50 PSI absolute. A rotometer and throttling valve to control air flow followed. The compressed air was cooled to 40° F. by a conventional Freon-12 refrigeration unit. This first cooling condensed most of the moisture, which in turn was removed by a water trap.

The air was then passed through a molecular sieve (Linde Corporation type 13X) which removed water to a dew point of −120° F. and lowered the carbon dioxide concentration to one part per million. Two such beds were used alternately on an 8-hour cycle, permitting heat regeneration of the spent bed.

The air temperature was finally reduced to the operating temperature of the charcoal column with a compound cascade refrigeration unit, using Freon-13 and Freon-22 as refrigerants, and three 7.5-horsepower compressors. The air was first cooled to −50° F. and then cooled to −125° F., the operating temperature of the charcoal bed.

The air was then passed through the radon absorption tower containing 30 pounds of 6–14 mesh activated coconut charcoal, where the radon was removed. The effluent air from the bed was passed through a rough filter, reheated to a thermostatically controlled comfort temperature, and passed through an ultrafilter to remove any charcoal that might have been carried over. Introduction of the air into the acclimation room completed the processing cycle. Reduction of ambient radon levels to the necessary acclimation room level required approximately 2 hours.

Collection System

The system for collecting the subject's breath consisted of a half-face mask, surge absorber, caustic spray tower, water freeze traps, charcoal columns, vacuum pump, and wet test meter (fig.

4). The mask, worn only during the collection of 15-minute breath samples, was located inside the acclimation room. The mask had a valving arrangement that permitted acclimation room air to be inhaled and the expired air to be exhaled through a 1-inch-diameter flexible tube which passed through the room wall to the collection system. If any leakage occurred at the face mask, a slightly lower pressure in the collection system than in the acclimation room pulled air into the system rather than letting part of the breath sample escape. The surge was a 5-liter rubber bellows, counterweighted to minimize pressure variations in the system.

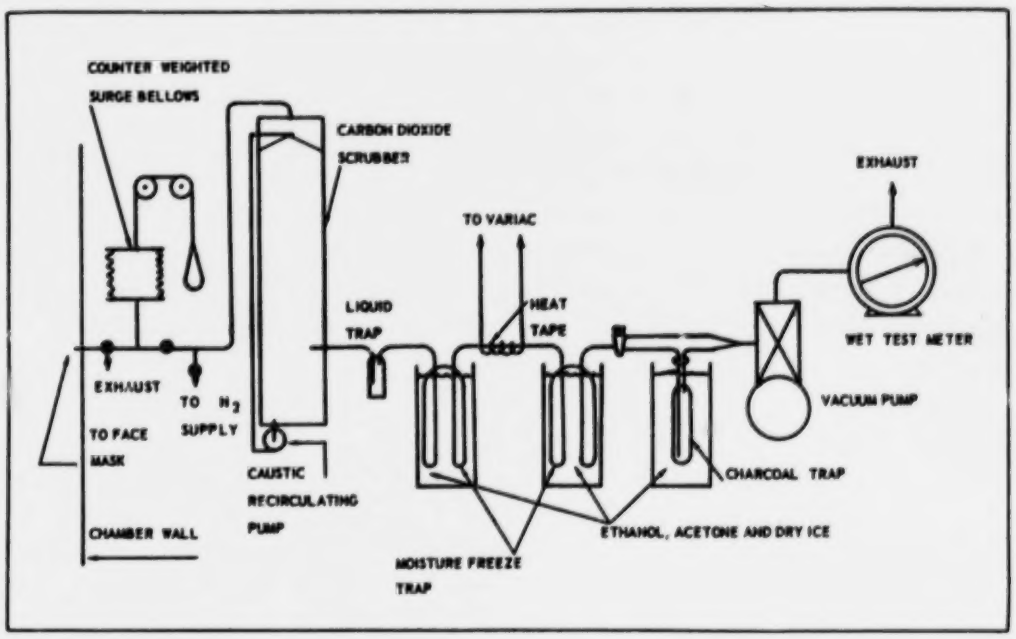

Figure 4. Radon Breath Collection System

The breath sample passed from the surge absorber to the caustic spray tower where the carbon dioxide concentration was lowered to 100 ppm. The caustic spray was housed within an 8-inch-diameter, 60-inch-high stainless steel cylinder. A centrifugal pump recirculated ½ gallon of 8*N* potassium hydroxide at a rate of 1 gallon per minute. The KOH was sprayed into the cylinder through a hollow cone nozzle. Following the CO_2 scrubber, a small liquid trap removed any KOH droplets or foam which may have been carried over.

Two freeze-out traps followed in line to condense moisture from the breath which would greatly inhibit collection of the radon if allowed to reach the charcoal. The moisture freeze traps

consisted of 1-inch-diameter glass tubing looped twice vertically into a 6-inch-diameter Dewar flask containing a solution of ethanol, acetone, and Dry Ice. It was found that 1 pound of ⅛-inch-diameter glass beads in the second loop assisted in the collection of ice crystals. Two such freeze-out traps were placed in series, with a heated copper tube between, measuring three-eights of an inch in diameter and 2 feet long. Most of the moisture was condensed in the first 4 inches of the first loop. The beads in the second loop acted as a physical stop for any ice crystals that may have been carried over. The heated cooper tube melted any ice crystals carried out of the second loop, and the entire process was repeated in the second freeze-out trap.

The charcoal radon trap was downstream from the moisture freeze-out traps. The radon trap was a flow-through column containing 3 to 5 grams of 12–14 mesh activated coconut charcoal. The charcoal trap was heated to approximately 700° F. 2½ hours before a breath sample was collected. The trap was kept at this temperature for 15 minutes while being alternately evacuated and flushed with aged nitrogen. This cycle was repeated three times and then the trap was sealed and allowed to cool under 10 PSI absolute nitrogen pressure to room temperature. The trap was then placed in a cold bath of ethanol, Dry Ice, and acetone, and connected in line with the rest of the collection system previously described. The charcoal radon trap was then ready for use.

After each breath sample was passed through the radon trap, acclimation room air was flushed through. This insured that none of the breath sample was left in the collection train. The radon trap was evacuated, sealed and replaced with a fresh trap. Each time the charcoal radon trap was replaced, the upstream moisture freeze trap was removed. The downstream moisture freeze trap was moved to the lead position, and a fresh, dry freeze trap was placed in the downstream position. This system was purged and checked for leaks after each change. The system was alternately purged with 30 liters of acclimation room air and evacuated three times prior to sampling. The breath sample, having passed through the radon trap, was drawn through the vacuum pump and exhausted to the atmosphere through a wet test meter. The pump, with a free air capacity of 140 liters per minute, was found to be barely adequate for this use because of the flow resistance of the system.

The wet test meter was used to measure the volume of air taken in samples and to measure the amount of acclimation room

air flushed through the system after the sampling. Actual measurement of the breath sample volume was not required since breath radon was calculated on a time-of-collection basis. A complete breath sampling cycle for two subjects required 3 hours. This included two air samples taken outside the trailer, two acclimation room air samples, and four breath samples.

Transfer System

The system for transferring radon from the charcoal trap to the counting cell was similar to the one described by Lucas (*9*). However, additional modifications and improvements were developed. Among these were an automated system to operate the repetitive cycles and various devices to reduce contamination of the scintillation cells. This system consisted of a heating unit, quick-connect O-ring joints, solenoid valves, compressed nitrogen tank, Sigmamotor pump, pressure gage, and an electrical controlling circuit. The unit controlling the repetitive cycles during the transfer was constructed from a synchronous motor, two plastic cams, and several microswitches. The pressure gage activated a relay that automatically shut off the unit when the transfer was completed.

After the radon was concentrated in the charcoal column, the column was removed and allowed to warm to room temperature. It was then connected to the transfer system (fig. 5) where it was heated to approximately 700° F. During the heating of the column, which lasted 30 minutes, the remainder of the transfer system was evacuated. The absence of leaks was established by observing the pressure gage. After the radon trap had reached the desired temperature, the system (excluding the radon trap and scintillation cell) was alternately evacuated and flushed with aged nitrogen. After the last evacuation, the outlet solenoid valve was closed, and glass valves located on the charcoal column and the scintillation cell were manually opened. Transfer began when the automatic recycling unit was turned on.

A full transfer cycle involved the following steps: (1) aged nitrogen was released into the heated and evacuated column; (2) the nitrogen supply was closed and the nitrogen remaining in the column was heated for a fixed time; and (3) the Sigmamotor pump then displaced the aged nitrogen and radon into the scin-

tillation cell. These steps were repeated until atmospheric pressure was reached in the cell. A complete transfer took approximately 20 minutes, at which time the glass valve on the scintillation cell was manually closed. Four hours later, after ingrowth of the radon daughters, the activity was counted in a scintillation counter.

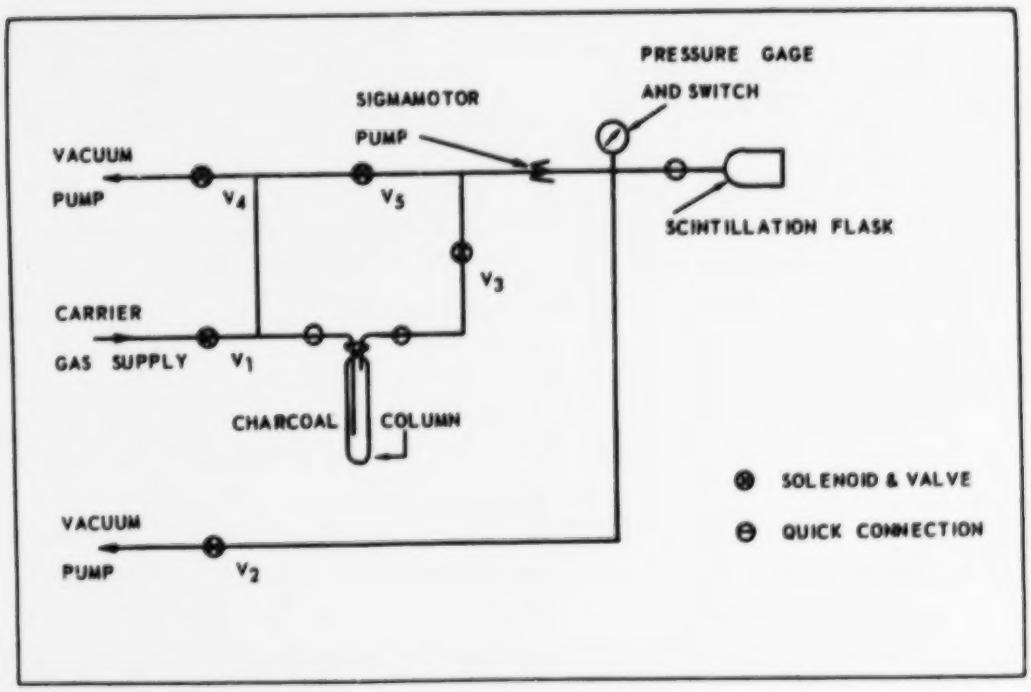

Figure 5. Radon transfer system

Counting System

After the scintillation cell was sealed, it was transferred to a specially constructed, low-background, alpha-scintillation counting system which was designed and fabricated at the Robert A. Taft Sanitary Engineering Center of the U.S. Public Health Service (*10*). The scintillation cells were basically Lucas-type counting cells made by following a procedure outlined by Rushing (*9*, *11*, *12*). Samples were counted for 15 to 20 hours. The entire counting system, including the scintillation flasks, was operated with backgrounds of less than 10 counts per hour. Although lower backgrounds could be obtained, less than 10 counts per hour were satisfactory for normal operations. A cell was counted each time on the same counter to provide the most consistent results obtainable.

BREATH SAMPLING

Two subjects who had eaten, bathed, and then dressed in freshly laundered clothing entered the acclimation room in the late afternoon or early evening. Drinking water, canned fruit juice, and a light snack were also placed in the room. Two hours prior to the subjects' entry, the air processing unit was started. Air leakage during the subjects' entry was assumed to have only a short-term effect on the radon concentration in the room.

After approximately 14 hours in the acclimation room, the subjects were awakened and permitted to drink a 6-ounce can of fruit juice. During this time, outside air samples and acclimation room air samples were taken. Within 1 hour of awakening, each subject alternately provided two 15-minute breath samples. The breath samples were collected with the subjects in a supine position (fig. 6).

Martin and Ferguson have investigated various aspects of the sampling procedure (*13–15*). They found no major or consistent

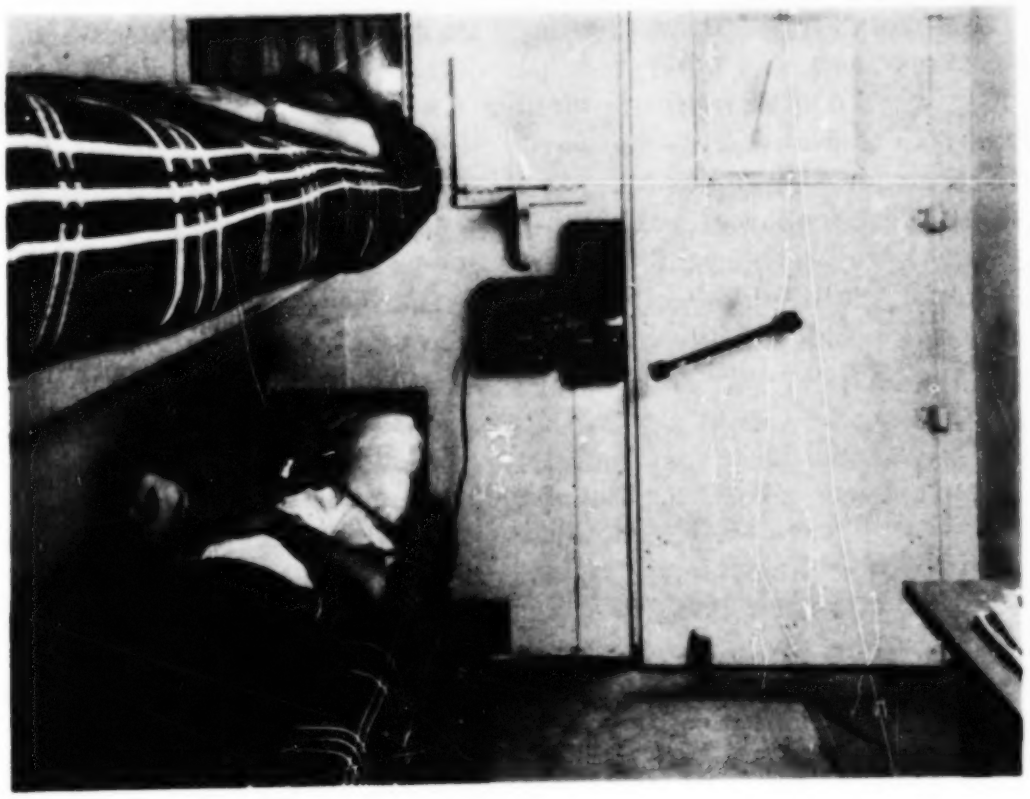

Figure 6. Sampling of subjects

effects on the radon content of expired air resulting from the feeding or the fasting of the subjects. They did find that the radon content of the expired air and the output of radon per unit time was much greater in the supine position than in the erect position, with the sitting position being intermediate. They also found that fluctuations in the radon content of the breath were smallest in the supine position after the subject had been in the position for 45 minutes.

Several investigators have found that 70 percent of the radon formed in the body is eliminated in the breath (*4*, *8*). Although this has been demonstrated only for radium body burdens between 10^{-5} and 10^{-8} gram, indications are that 70 percent is a minimum figure (*8*).

EVALUATION OF PERFORMANCE

Immediately prior to making breath radon measurements on area residents and laboratory personnel, 14 radon standards (4.8 pCi) were run through the double freeze-out traps and onto the charcoal columns. Nine standards (4.8 pCi) were run through the KOH tower. The recoveries and associated 95-percent-confidence levels were 81.80 percent ±5.34 percent and 82.18 percent ±7.28 percent, respectively. The final step was to run two standards through the total collection system from the room wall through the bellows, KOH tower, and freeze-out traps to the charcoal. Recovery for the two standards was 80.63 percent and 81.67 percent.

The investigators felt that these values were satisfactory and began a series of breath radon measurements on laboratory personnel and area residents. These measurements were halted 3 months later because of the termination of the San Juan Basin Radium Research Project.

The following results are presented only as an indication of the system's present capabilities. The body burdens cited in table 1 are in no way proffered as absolute values.

Low-level radon standards (0.48 pCi) were run through the total collection system for some of the series of breath radon measurements to act as a system performance check. When obvi-

Table 1. Radium 226 body burdens [a]

Subject [b]	Number of samples	Radium 226 body burden	
		Low (pCi)	High (pCi)
A	8	0	202
B	7	55	208
C	6	16	212
D	8	102	290
1	2	64	68
2	2	81	229
3	2	23	60
4	2	78	81
5	2	5	122
6	2	16	140
7	2	149	360
8	2	96	193
9	2	108	152
10	2	43	114
11	2	142	194
12	2	104	133
13	2	108	205

[a]Based on 65.2 percent recovery and 70 percent exhalation factor.

[b]Subjects denoted by letters were laboratory personnel. Those denoted by numbers were area residents.

ous malfunctions in the equipment took place, the associated data were discarded.

The average recovery of 11 standards (0.48 pCi) was 65.2 percent with a 2-sigma-confidence interval of ±41.2 percent. For three standards run on the same day, the average recovery was 80.5 percent ±29 percent. Standards were not always run in conjunction with a particular breath radon measurement; therefore the average of the 11 standards (65.2 percent) was used to calculate the radium body burdens shown in table 1. Since the minimum radon recovery to be expected within the 2-sigma-confidence interval is 24.0 percent, the amount of radon (and therefore radium body burdens) might occasionally be four times the amount measured in the sample.

The investigators attribute the large variation in radon recovery to the collection system, particularly the rubber bellows and KOH tower. Modifications, to correct this source of error, were still being made when the project terminated.

Figure 7 shows the sources of radon in the breath for one of the laboratory personnel as a function of time in the acclimation room. The theoretical curve shown in figure 1 was constructed in

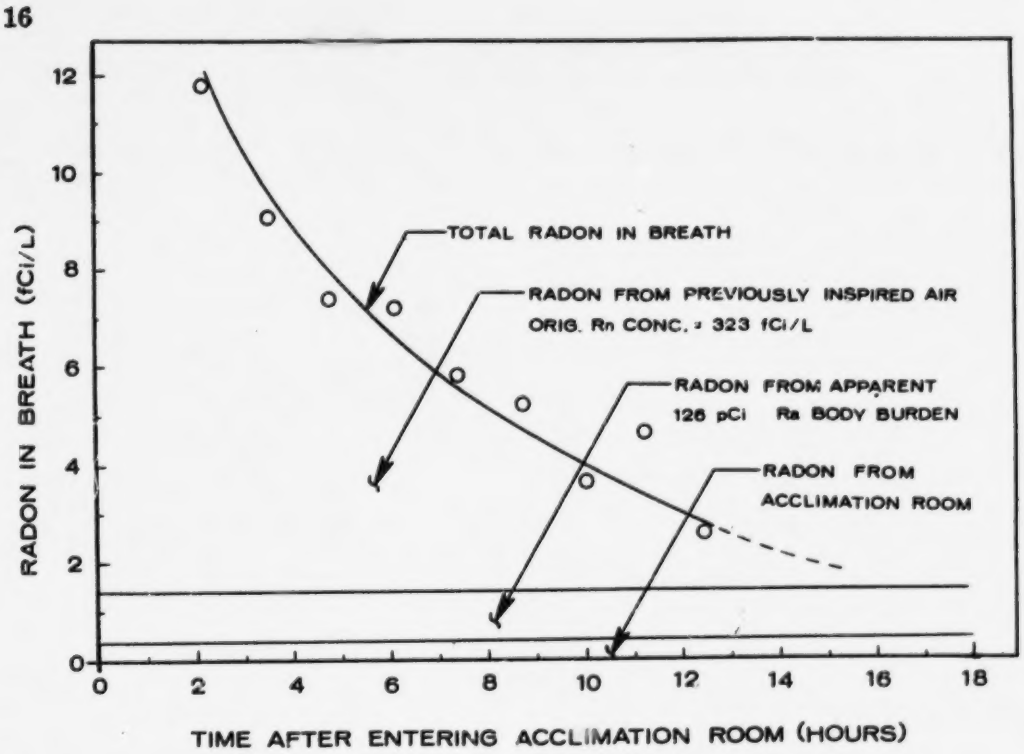

Figure 7. Sources of radon in breath (experimental)

such a manner that a direct comparison with figure 7 is possible. It can be seen that the previously inspired radon is exhaled at a slower rate than the theoretical curve predicts. It is not possible to determine if the elimination curve is or would be asymptotic with the average value obtained for the subject after 13 to 15 hours acclimation because of fluctuations in the data obtained from this subject.

On the basis of this curve alone, 15-hour acclimation periods do not appear to be sufficient time for the elimination of radon in the soft tissue. If this is true for the other subjects tested, their apparent radium 226 body burdens are higher than their true body burdens by an amount equal to the transient radon remaining in the soft tissue.

The average reduction of radon in the processed air was 600:1, based on 30 acclimation room air samples and 19 outside air samples. The maximum reduction achieved for any one day was 2,000:1, while the minimum for any one day was 150:1. These figures are based on the 65.2-percent average recovery. It can therefore be seen that the design objective of a hundredfold reduction was achieved. Radon concentrations in the acclimation room air were low enough that they did not obscure the radon from the radium 226 body burdens.

SUMMARY

The air processing unit reduced the radon concentration in the acclimation room from approximately 266 fCi/liter to 0.5 fCi/liter, thereby allowing a more accurate determination of low-radium body burdens. At the same time, the acclimation room provided comfortable surroundings for subjects during the extended acclimation periods required. Mobility of the entire breath radon apparatus was achieved.

Variability in sample recovery, attributed to operational difficulties in the collection system, appeared to be the limiting factor in the accurate determination of radium body burdens at these low levels. A complete evaluation and subsequent refinement of the collection system were not possible because of the termination of the San Juan Basin Radium Research Project. For the same reason, it was not possible to make a detailed study of the rate of elimination of radon from the body. Thus acclimation times, which would minimize the effect of previously inhaled atmospheric radon, were not determined. Satisfactory operation of the air processing unit and subject cooperation in using the acclimation room indicate that this unit would be particularly suitable for this purpose. The mobility of the breath radon unit further enhances this possibility.

Before the full import of the breath radon apparatus can be realized as a research tool for the measurement of radium body burdens, an evaluation of the factors mentioned previously is necessary.

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U.S. Department of Health, Education, and Welfare, Public Health Service Publication No. 999–RH–31 (1968), 19 pp. (limited distribution).

ABSTRACT: A mobile breath radon detection apparatus has been used for measuring low radium 226 body burdens. The unique feature of the apparatus is an acclimation room designed to accommodate subjects for periods up to 1 week in a low-radon environment. Radon concentrations in the acclimation room have been reduced to an operating level of 0.0005 pCi per liter, a reduction of approximately 600:1, by an air processing unit employing activated coconut charcoal. This apparatus permits determination of very low radium 226 body burdens in comfortable surroundings for two subjects simultaneously, during the extended periods required for elimination of previously inhaled radon from the body. The construction and operation of the acclimation room and breath sample collection are described. Radium 226 body burdens for the subjects and a radon elimination curve for one subject are included.

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